

Waste Printed Circuit Board (PCB) Recycling Techniques

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Abstract With the development of technologies and the change of consumer attitudes, the amount of waste electrical and electronic equipment (WEEE) is increasing annually. As the core part of WEEE, the waste printed circuit board (WPCB) is a dangerous waste but at the same time a rich resource for various kinds of materials. In this work, various WPCB treatment methods as well as WPCB recycling techniques divided into direct treatment (landfill and incineration), primitive recycling technology (pyrometallurgy, hydrometallurgy, biometallurgy and primitive full recovery of NMF-non metallic fraction), and advanced recycling technology (mechanical separation, direct use and modification of NMF) are reviewed and analyzed based on their advantages and disadvantages. Also, the evaluation criteria are discussed including economic, environmental, and gate-to-market ability. This review indicates the future research direction of WPCB recycling should focus on a combination of several techniques or in series recycling to maximize the benefits of process.

Keywords Waste printed circuit board · Recycling techniques · Pyrometallurgy · Hydrometallurgy · Mechanic separation · Performance evaluation

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Abbreviations

EEE	Electrical and electronic equipment
WEEE	Waste electrical and electronic equipment
PCB	Printed circuit board
WPCB	Waste printed circuit board
MF	Metallic fraction of printed circuit board or waste printed circuit board
NMF	Non-metallic fraction of printed circuit board or waste printed circuit board

1 Introduction

Electrical and electronic equipment items (EEE) including MP3, cell phones, and tablets have been the indispensable necessity of peoples' daily lives while past decades have witnessed the rapid growth of EEE manufacturing industries. Currently, the production rate of EEE is estimated to be between 8.3 and 9.1 million tons per year in 2005 with an increasingly rapid growth rate due to technology updates, making it the dominant manufacturing industry in the world [1, 2]. The rapid development of EEE has given rise to the blooming of the printed circuit board manufacturing industry, which produces the core components of electrical and electronic equipment [3, 4]. Meanwhile, the accelerating upgrading of these industries also shortens the life span of EEE from around 5 years to as short as 2 or 3 years [5, 6]. This is an accelerating process, with an annual rate of 3–5% according to an EU report in 2000 [7]. Consequently, it is not difficult to realize the fact that the amount of waste electrical and electronic equipment (WEEE), as the result of EEE jettisoning, is increasing at an overwhelming rate. It is estimated that currently the amount of WEEE production is nearly 45 million tons yearly worldwide with an annual growth rate of 7–10% [8].

The printed circuit board (PCB) is considered to be the core part in most kinds of EEE (see Table 1). It is estimated that global production of printed circuit boards (PCBs) was around \$50 billion in 2010 and reached nearly \$60 billion in 2012 with a market growth rate of 1.7% [10, 11]. However, the mass disposal of WEEE generates a huge stream of waste printed circuited board (WPCB) into municipal solid waste, which was produced in large amounts which increase significantly every year (Fig. 1). Although WPCB only occupies a small portion around 3–6% of

Table 1 Main components of typical WEEE [9]

Typical WEEE	Main components
Refrigerator	Tubes, liners, condenser, wires, refrigerant
Air conditioner	Heat exchanger, motor, compressor, copper pipe, PCBs, wires, refrigerant
Washer	Tub, drain hose, motor, wires, salt waste
Television	Deflection yoke, demagnetized coil, speaker, PCBs, wires, CRT, LCD
Computer	Speaker, battery, storage medium, PCBs, wires, CRT, LCD
Printer/duplicator	Roller, toner, PCBs, wires, toner cartridge

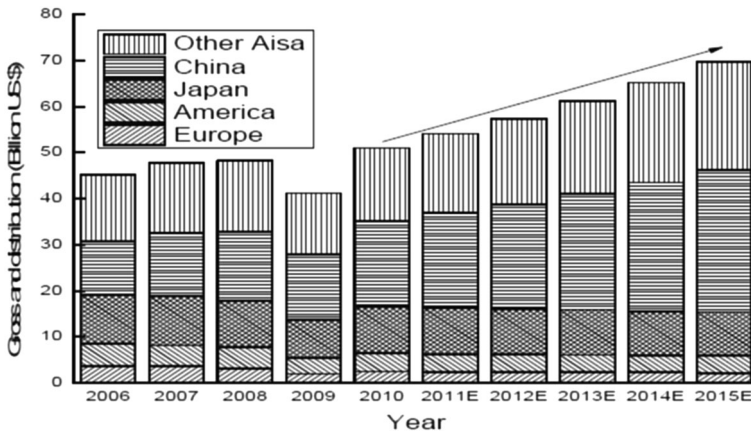


Fig. 1 PCBs increasing based on the global gross [11]

the total WEEE generated worldwide, the immense total WEEE amount still made it considerable [8, 12]. WPCB can be either directly treated, which includes landfill and incineration, or the recycling of WPCB by various kinds of technologies is also an alternative [13–18]. Compared to direct treatments, recycling is more favorable due to both environmental and economical consideration given the enrichment of materials in WPCB. It is regarded as a secondary resource since the concentration of precious metals, organic resins, or polymers in WPCB is normally ten times higher than rich non-renewable are (sometimes even hundred times higher) [19]. Therefore, recycling is not simply reduction of waste but the reuse of resources with better economic feasibility and less environmental impact.

The USEPA (United States Environmental Protection Agency) has identified seven major benefits when scrap iron and steel are used instead of virgin materials (Table 2). Using recycled materials in place of virgin materials results in significant energy savings (see Table 3 [20]). Also, the European Commission launched the 2002/96/EC Directive, known as the WEEE Directive, which came into effect on February 13, 2003, with the aim of achieving up to 70–80% recovery of electrical and electronic equipment [15]. Therefore, recycling is not only for the purpose of resource refining, more importantly, it can reduce the toxic pollution emitted in the initial manufacturing process, which will make a significant benefit difference in remediation costs.

Current recycling methods includes pyrometallurgy and hydrometallurgy, which have a long history and wide application [21–23]. Biometallurgy as an emerging technology also takes up a certain share of the WPCB recycling market [24–26]. Many studies have been done to improve the performance or cut the cost of these techniques. However, the inherent drawbacks are very obvious for these techniques, including the irritation to the environmental intensive energy consumption. Therefore, new advanced technologies are in great demand due to the requirement of technology with high safety and economic feasibility. Mechanical separation was developed due to the requirement for a high-purity metallic fraction of WPCB (MF)

Table 2 Recycled materials energy savings over virgin materials

Materials	Energy savings (%)
Aluminum	95
Copper	85
Iron and steel	74
Lead	65
Zinc	60
Paper	64
Plastics	>80

Table 3 Benefits of using scrap iron and steel

Benefits	Percentage
Saving in energy	74
Saving in virgin materials use	90
Reduction in air pollution	86
Reduction in water use	40
Reduction in water pollution	76
Reduction in mining wastes	97
Reduction in consumer wastes generated	105

and it can achieve good separation performance for real-use applications [27–30]. Recently, the use of the non-metallic fraction (NMF), which was forgotten in the past, is becoming the concern of researches including both the direct use or the modification of NMF [31–35].

However, a unanimous blind spot among the majority of works is the environmental emission analysis, which is a very serious problem derived from WPCB treatment. Several studies have been done on the evaluation of the emissions or discharge [5, 15]. Common pollutants generated from WPCB treatment or recycling include heavy metals, secondary particulates, PBDD/Fs, and PCDD/Fs, which have been studied and their toxicities are regarded as obstacles to WPCB recycling. However, emissions with lower toxicity, especially bromide in the form of hydrogen bromide or bromide, did not receive adequate attention. Despite the relatively low harm these emissions possess compared to dioxin-like organic compounds or heavy metals, the jeopardy they show is still a significant threat to human health that requires detailed studies and further treatment. Moreover, the complexity of WPCB's composition creates the scenario that specific analysis targeting certain pollutants is available while an overall review of emissions impact is still absent. Therefore, this overall emission impact study and strategy needs more effort.

Other than the discussion of emission, another issue mentioned previously is concerned with the techniques that focus mainly on general WEEE treatment methods. Nevertheless, WPCB shares several similarities with WEEE in terms of composition and chemical characteristics, the minor differences between them still requiring the development of better process techniques for WPCB treatment [36–38]. The common composition of EEE is shown in Table 4 while the detailed

Table 4 Main materials found in EEE [39]

Material	Percentage
Ferrous	98
Non-ferrous	28
Plastics	19
Glass	4
Wood	1
Other	10

discussion of WPCB will be presented in a later section. Therefore, the techniques for WEEE can be applied to WPCB but may not acquire ultimate efficiency. Actually, compared with WEEE, the composition of WPCB is more or less more consistent, since the outer casing for WEEE varies with different kinds of EEE, for example, the refrigerator or television while most WPCB has no relationship with the outer casing, making WPCB simpler for treatment or recycling. Thus, the specific design and analysis for WPCB treatment or recycling is indispensable in order to obtain the maximum sustainable environmental and economical benefits.

Herein, the lack of a summary on current techniques due to the upgrading of techniques as well as the absence of systematic emission analysis claims that an overall review of this field is imperative. Also, with the development of technologies, the overlapping of chemical, mechanical and physical treatments is very obvious, thus a new criteria is necessary for sorting and evaluating the interdisciplinary recycling and treatment techniques. In this work, the treatment and recycling techniques of WPCB are categorized according to their recovery rate and both existing and emerging techniques are reviewed and discussed. The performance of WPCB recycling techniques is evaluated from both an economic and environmental perspective. Moreover, the concept of “gate to market ability” is introduced for a more profound understanding of the feasibility of WPCB recycling techniques.

2 Characterization of WPCB

The composition of WPCB varies, since the source, components, and manufacturing processes of the WPCB maybe very different [40]. Generally, the composition of currently used PCBs is classified as glass fiber reinforced epoxyresin (FR-4), which is normally used for small electronic equipment or a paper laminated phenol is resin (FR-2) used in home appliances [41, 42]. Also, during the manufacturing process, carbonaceous compounds including brominated flame retardants and dyes are added in order to improve the flame resistance and visual recognition ability. In Table 5, it is obvious that WPCB mainly consists of MF and NMF. It is generally accepted that WPCB contains around 30% metallic fraction and 70% non-metallic fraction by weight [43, 44]. However, what needs to be clarified is that the percentage of noble metals in WPCB has been decreasing in recent years with the upgrading of technologies [45].

It is widely accepted that WPCB is comprised of a heterogeneous mixture [39, 46–48]. The heterogeneity originates from two aspects, which are the

Table 5 Composition of a populated PCB [36]

Components of WPCB	Percentage
Non-metal (e.g., epoxy resin, glass, etc.)	>70
Copper	~16
Solder	~4
Iron, ferrite (from transformer cores)	~3
Nickel	~2
Silver	0.05
Gold	0.03
Palladium	0.01
Other (bismuth, antimony, tantalum, etc.)	<0.01

heterogeneity of different PCB brands including the use of cellulose paper reinforced phenolic resin or glass fiber reinforced epoxy resin and heterogeneity in the morphology of a PCB due to the presence of metal plating [49]. However, if an adequate amount of WPCB is collected and pulverized into fine powder, empirically rules of the composition of WPCB can still be identified by comparing them globally. This is the so-called “pseudo-homogenous” effect, which means that the individual composition of a certain WPCB item will vary from others, while the bulk composition of all WPCB items will still share similarities, as shown in Table 6.

Generally, the uncertainty of WPCB composition hampers the systematic treatment or recycling of WPCB, since the complexity of it interferes with each component [59]. Therefore, pulverization is a very important pretreatment part for mixed WPCB recycling and treatment techniques since the higher pseudo-homogeneity is more favorable for easier application in overall treatment.

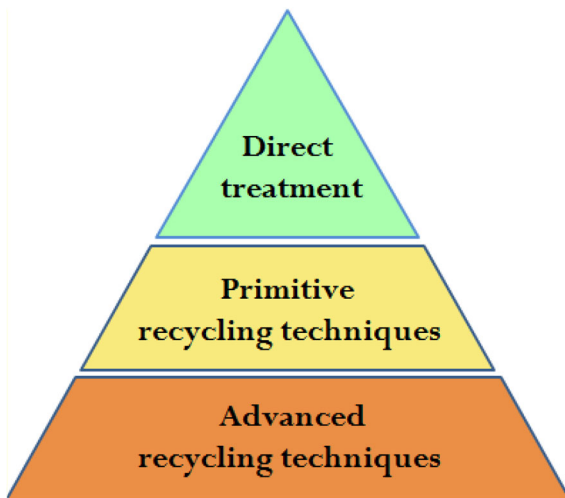
3 Direct treatment methods—landfill and incineration

The types of technologies for WPCB recycling are increasing with the development of research and thus the old category for technologies sorting is no longer applicable. Some processes divide the technologies to thermal or non-thermal, or

Table 6 Materials composition of several common used printed circuit boards

Metals/Ref.	[15]	[50]	[51]	[52]	[53]	[54]	[55]	[56]	[57]	[58]
Cu/%	36.4	16.0	33.87	18.29	19.187	32.5	19.60	31.95	26.8	19.19
Au/%	–	0.025	0.0007	0.011	0.013	0.004	–	–	0.008	0.007
Ag/%	–	0.100	0.04	0.018	0.0704	0.3	0.0034	0.34	0.33	0.01
Al/%	8.7	5.0	2.57	–	4.011	3.7	2.34	11.11	4.7	7.06
Pb/%	3.8	2.0	4.96	2.31	0.385	–	0.30	2.41	–	1.01
Pd/%	–	0.010	0.0012	–	0.0028	–	0.0016	–	–	–
Sn/%	4.6	3.0	0.153	3.92	0.689	0.9	0.17	1.78	1.0	2.03
Cd/%	–	–	0.00002	–	–	–	–	–	0.015	–
Total	53.5	33.14	41.59	24.55	24.36	37.40	22.42	47.59	32.85	29.31

Fig. 2 Hierarchy figure of the WPCB treatment and recycling techniques



some divide into chemical treatment or mechanical treatment, others divide them by the atmosphere of the reaction or some divide them into hydrometallurgical processing or pyrometallurgical processing [16, 60, 61]. In this review, the treatment and recycling techniques for WPCB are classified according to their recovery degree (Fig. 2). Direct treatment including landfilling and incineration means that no or only the energy of WPCB was recovered while primitive recycling means the simple recycling of MF, while the NMF ended up in disposal or limited non-hazardous treatment. Advanced recycling includes two parts: separation methods without damage to the NMF and the direct use or modification of NMF. It is highly convincing that this classification can better describe the current situation of WPCB recycling and provide a basis for an overall treatment perspective instead of focusing on only one or two areas.

3.1 Landfill

Landfill is a WPCB treatment method with a long history and wide application worldwide because of simplicity in operation. However, the land for WPCB disposal is thereby regarded as wasteland and normally incapable of being exploited again in predictable decades, which is not suitable for countries/communities where there is a lack of land space. In addition, the environmental issues aroused by WPCB landfilling such as the leachate formation in landfill sites and evaporation of hazardous substances raise safety concerns. It is reported that almost 70% of the heavy metal in landfill sites come directly from WEEE, which indicates that a considerable part of heavy metals actually come from WPCB [62]. Concurrently, the co-landfilling of WPCB with various kinds of municipal solid waste makes the formation of leachate containing brominated toxic compounds and heavy metals possible due to the reactions during the landfilling process [13, 14]. The synergic pollution will contaminate the atmosphere, soil, and ground water, and this has been proven by several research groups.

Heavy metals including lead and mercury are identified in landfill sites. Lindberg et al. identified total gaseous mercury (TGM), monomethyl mercury (MMM), and dimethyl mercury (DMM) in the landfill gas in Florida, USA [63]. The results show that the mean concentration of TGM is 7190 ng/m^3 and MMM is 6 ng/m^3 . It should be emphasized that DMM, a form of extremely toxic organic mercury, was also observed in landfill gas with a concentration of 30 ng/m^3 . Spalvins et al. also identified lead in the leachate in landfill sites with a concentration of $0.007\text{--}0.066 \text{ mg/l}$ [64]. Other than heavy metals, PCDE(polychlorinated diphenyl ethers)/PBDE(polybrominated diphenyl ethers) were also discovered in landfill sites by Osako et al. [65]. Samples from seven different landfill sites in Japan were collected and analyzed. Landfill sites in operation or closure within 1 year show the presence of several kinds of hazardous compounds, especially obvious is the higher concentration of brominated flame retardants and the derivatives. TBBPA with concentration from n.d. to $620,000 \text{ pg/l}$ and PBDE-47, -99, and -100 with the total concentration from n.d. to 4000 pg/l were detected in the raw leachate from different landfill sites. More dangerously, the hazards of landfill leachate and gas are not only limited to the landfill site because they will propagate through aquatic systems or rain fall and cause the spread of contamination. Wong et al. found that even in the downstream of the small-scale landfill site of a Chinese processing village, a certain amount of metals can still be detected including Cd (n.d. to 10.3 mg/kg), Cu ($17.0\text{--}4540 \text{ mg/kg}$), Ni ($12.4\text{--}543 \text{ mg/kg}$), Pb ($28.6\text{--}590 \text{ mg/kg}$), and Zn ($51.3\text{--}324 \text{ mg/kg}$) [66]. This indicates the possibility of toxic substance migration, which makes the condition of landfill sites much worse.

In summary, landfilling is currently considered an improper way to treat WPCB due to the environmental concerns it brings about, and actually the function of it is to use land resources to remedy hazardous WPCB, which is a waste of resources for both soil and WPCB. Therefore, stricter policies and regulations have been imposed on simply landfill WPCB in many countries and more environmental benign treatment methods are required [67–71].

3.2 Incineration

Incineration means the combustion of WPCB by converting its calorific value to energy and emitting the gas directly or after treatment with the purpose mainly to remove the non-metallic fraction part (around 70 wt%) [72, 73]. It has the advantages of significantly reducing the volume of WPCB by 50%, and also the calorific value of WPCB is relatively high compared to municipal solid waste, which is around $9.9 \times 10^4 \text{ kJ/kg}$ [15, 16]. Therefore, it readily satisfies the minimum incineration calorific value for waste, which is roughly 5000 kJ/kg . Currently in the world, incineration is still widely used in American, Asia, and Europe due to the simplicity of the process. However, during the combustion of WPCB, toxic emissions including heavy metals, fly ash, polychlorinated dibenzo-*p*-dioxins/polychlorinated dibenzofurans (PCDD/Fs) and polybrominated dibenzo-*p*-dioxins and dibenzofurans (PBDD/Fs) are released into the atmosphere in the absence of post-purification [74, 75]. Cadmium, copper, nickel, lead, and zinc will be vaporized according to the order of their melting points and released into the

atmosphere [76]. Unfortunately, incineration is an ideal place for the formation of PCDD/Fs and PBDD/Fs due to the presence of halogens and an oxidizing atmosphere as well as incomplete combustion [77–79]. Additionally, in incineration, copper in WPCB will play the role of catalyst for dioxin formation in the presence of brominated flame retardant [5]. This all contributed to the high emission amount of PBDD/Fs, as high as 130,000 ng/g, which is very high compared to other investigations [80]. Also, the amount of PBDD/Fs exceeds PCDD/Fs significantly due to the abundance of bromide in WPCB. Other gaseous pollutants including Br₂, HBr, CO, and NO_x also attract considerable concern about them.

Ni et al. has conducted high-temperature WPCB incineration in a tube furnace and found that high-temperature combustion favors the removal of PBDD/Fs, which can release maximum 99.9% bromide into flue gas in the form of HBr or Br₂ with much lower toxicity compared with PBDD/Fs [79]. Also, the temperature needs to exceed 1200 °C in order to reduce the formation of CO and ensure the completely decomposition of brominated flame retardant (Fig. 3). However, the high temperature will slightly favor the formation of NO_x from 690 mg/Nm³ (800 °C) to 790 mg/Nm³ (1400 °C) that exceeds the emission standard value of 500 mg/Nm³. In spite of HBr and Br₂ having a lower toxicity, they still need further treatment like adsorption to avoid the emission to local atmosphere.

Other than the gaseous emission that incineration releases, there is still the solid-phase residue of the incineration that is termed “bottom ash,” which will be landfilled or processed for further treatment. There are significantly enriched heavy metals in the bottom ash including copper (35.8–1295 mg/kg), Pb (226.3–606.6 mg/kg), and Cd (0.87–7.46 mg/kg), which makes the safe disposal of it become a troublesome issue [76]. Normally, a vitrification process at a temperature of 1500–1600 °C is typical for the treatment of bottom ash [82].

Therefore, incineration as a treatment method for WPCB is not an environmentally friendly option considering the toxic emissions. Also, the construction of an incineration plant is a major expenditure for local governments. Actually, as these side effects were revealed by the emerging researches, the use of existing incineration plant or planning for future incineration plants was seriously affected in many countries. Public concerns and acceptability were more attracted to the recycling instead of simple treatment of WPCB.

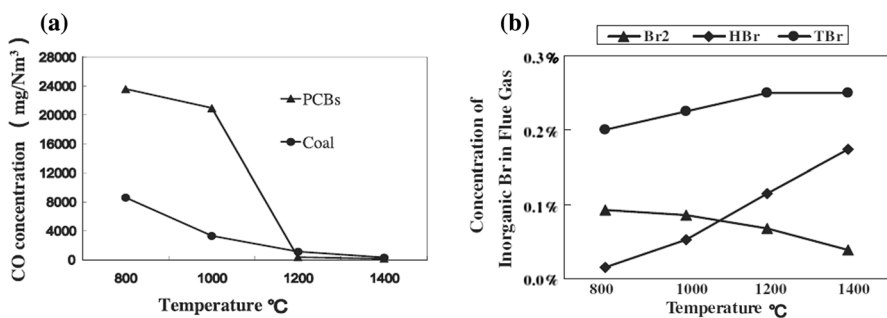


Fig. 3 Effects of temperature on CO emission (a) and HBr/Br₂ emission (b) (excess air factor = 1.3, high temperature zone residence time = 0.25 s)

4 Primitive Recycling Techniques

4.1 Partial Recovery of Metallic Fraction from WPCB

4.1.1 Pyrometallurgy Recycling Techniques

Pyrometallurgy recycling techniques are conducted by decomposing WPCB in the absence of air or the presence of an inert atmosphere. It is a very important and mature technology for WPCB recycling and numerous works have been done on this. Although pyrometallurgy shares similarities with incineration, they are still distinct and separate techniques. The main difference between pyrometallurgy and incineration is that the purpose of incineration is simply reducing the volume of WPCB and to, achieve partial heat recovery though the incineration of WPCB when the heat released is adequate, whereas the purpose of pyrometallurgy focuses more on resource recovery, which includes the concentrating of MF and other materials out of the WPCB feedstock by decomposing NMF to low molecular weight organic compound-formed liquid or gas. This also leads to the result that unlike incineration, the operation cost for pyrometallurgy is normally much higher than incineration due to maintaining the absence of oxygen from the process. More comparisons of incineration and pyrolysis in detail are listed in Table 7.

In pyrometallurgy, the process is typically divided into two stages. In the first stage, the WPCB starts to decompose due to the intensive heat input and the release of volatile organic compounds. The second stage is the formation of char due to the pyrolysis of polymer inside the structure [6]. Therefore, temperature is a very critical variable in pyrometallurgy such that low temperatures (<400 °C) will favor the formation of liquid products while high temperatures (>800 °C) will help the breakage of high molecular weight compounds, thus producing more small organic molecular [15, 83, 84]. Also, the pyrolysis of WPCB is extremely dangerous while the temperature is low (lower than 800 °C) and the absence of an inert atmosphere since there will be PBDD/Fs formation [85]. Normally, the absence of air is relatively difficult to ensure, especially in large-scale applications, thus the production of PBDD/Fs is a serious issue for low-temperature pyrolysis.

As for the pyrolysis products, some pyrolysis reactions can produce pyro-oil or pyro-gas, which has the potential to be used as fuel or chemical material source, but the complexity and uncertainty of it greatly hampers its application [15, 16]. Also, the products of pyrometallurgy have been characterized by several groups and HBr, by-products of brominated phenols, PCDD/Fs, PBDD/Fs have been found in pyrolysis products, which has the side effects of aquatic toxicity, carcinogenicity, and mutagenicity [17, 18, 86–89]. As for the pyro-solid, which is the inorganic components left after pyrolysis normally generates a huge amount of acidic effluents, will jeopardize the aquatic environment and cause side effects to human health [90]. As a result, the majority of research only focuses on the recovery rate of WPCB without overall consideration of the environmental impact analysis.

Pyrolysis of commercial WPCB has been done in an autoclave at 500 °C for 30 min and the pyro-liquid, pyro-gas, as well as pyro-solid were collected [15]. The

Table 7 Comparison between incineration and pyrolysis [81]

Aspect	Incineration	Pyrolysis
Dioxins and furans emissions	Incineration produces dioxins that are released in air and remain also in the solid material	Pyrolysis has a reduced production of dioxins
Bromine recovery	Incineration achieves complete bromine evolution in combustion gases. There is a risk of formation of elementary Br ₂	Bromine evolves part in the gas and part in pyrolysis oil. Bromine recovery could be an important aspect for the pyrolysis plant
Tin recovery	With incineration tin solder recovery is not possible	Tin solder recovery is possible with pyrolysis
Heat recovery	Incineration will release more heat than pyrolysis	Pyrolysis products will have 70 and 40%, respectively, of the raw material heating value (respectively for RAMs and CPUs)
Heavy metals emissions	Incineration results in the emission of mercury and cadmium	Pyrolysis process has reduced emissions of heavy metals

yields of pyro-liquid, pyro-gas, pyro-solid, and char derived from the pyrolysis of polymeric materials were 16.2, 7.3, 76.5, and 5.8%, respectively. However, the value of these pyrolysis products is very limited since the pyro-solid contains a high content of heavy metals including Cu (36.4 wt%) and Pb (3.8 wt%), which is not capable of being recovered and actually requires further treatment. Also, the pyro-liquid contains a certain amount of Br and Cl, which is hazardous to human health, and is not clarified in the paper. Guan et al. also studied the pyrolysis products of WPCB and the GCV value of pyro-liquid was found to be around 29 MJ/kg, which is around the same level as coal, which is normally 30 MJ/kg [16]. However, the Br inside the pyro-liquid will hamper the usage of it, which must undergo removal before application. The main components of pyro-gas are hydrogen, carbon monoxide, carbon dioxide, and methane at 500 °C. However, there still are some bromorganic molecules inside, which will affect the usage of the pyro-gas. Therefore, we can conclude that the failure of separation with Br is a serious challenge for the application of pyro-liquid and pyro-gas. Thus, Terakado et al. used metal oxide including ZnO, Fe₂O₃, La₂O₃, CaO, and CuO as the fixation of Br in pyrolysis process and the results show a significant reduction of HBr as well as bromorganic compounds emission [91]. The deficiency is that metal oxide was used as additive and still in the end bromide salt requires further treatment.

Guo et al. has done the pyrolysis in 500 °C and 71.60 wt% solid residue, 18.23 wt% tar, and 10.71 wt% gas were obtained from this process [92]. The main component of the pyro-gas is CO, CO₂, propylene, and small bromorganic molecules like bromomethane and bromopropane. The calorific value of pyro-gas is 2.386 MJ/kg, which is quite low comparing to natural gas (38.93 MJ/kg), together with the difficulty in direct application due to bromide inside the gas. The pyro-tar and pyro-solid have been investigated and been found to have a certain amount of metal and bromide, reducing its environmental friendliness. The energy balance was

calculated and it shows that only 35% of the heat required for pyrolysis is capable of recovery from the pyrolysis products, which means an additional heat source is necessary for the process.

Cayumil et al. has done high temperature pyrolysis from 800 to 1350 °C [93]. After this process, they obtained a slag phase, tin–lead (~40% Sn, ~8% lead, and ~45% copper)-rich metallic phase and copper-rich phase (~85% copper), which has relatively high purity. However, there are still 22–25 wt% of slag phase which has not be recycling and the crucial point is that there is still certain percentage of metal inside this slag phase, which means that although this process is very energy-intensive, there is still a considerable amount of toxic solid waste produced (Fig. 4).

To remedy the drawbacks of a high pyrolysis temperature and side reaction occurrence, vacuum pyrometallurgy techniques have recently been developed as an advanced technology for pyrometallurgy recycling techniques, since it has the advantages of low operation temperature and a reduction in the intensity and occurrence of side reaction or reformation of hazardous materials [94–96]. However, the strict vacuum requirement calls for a separation before vacuum metallurgical methods.

Anjan Kumari et al. conducted vacuum pyrometallurgy followed by solvent extraction to recycle various kinds of product after vacuum pyrolysis at 300 °C for 4 min due to the enhanced performance of pyrolysis under high vacuum [88]. This is a very systematic process that has the advantage to recycle fuel oil, fuel gas, sulfuric acid, Cu, Ni, and Fe. Therefore, the emission of toxic gas will be reduced significantly since the recovery section takes a big percentage of the total composition of WPCB feedstock. Details about this process are shown in Fig. 5. However, attention to the bromide is still not be given, since the process does not contain a step for bromide removal and TEHA dissolved in kerosene is used as the extraction solvent, which will also have the side effect of emission.

Zhou et al. found that the assistance of pre-separation will improve the efficiency of the process [49, 97]. The pretreatment is mainly aimed at separate solder and other electronic components. They performed the vacuum pyrolysis at 600 °C for 30 min under a vacuum lower than 1.5 kPa. However, the pretreatment was conducted at 183 °C, which needs an energy input as they are using an oil bath, which is not comparable to the result of separation of solder although the composition of solder remains almost unchanged. For the pyrolysis part, the pyro-

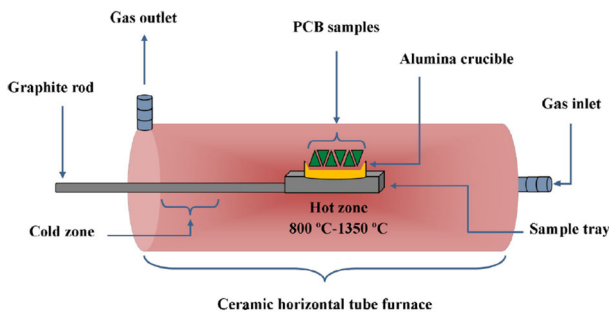


Fig. 4 Typical design of pyrolysis equipment [91]

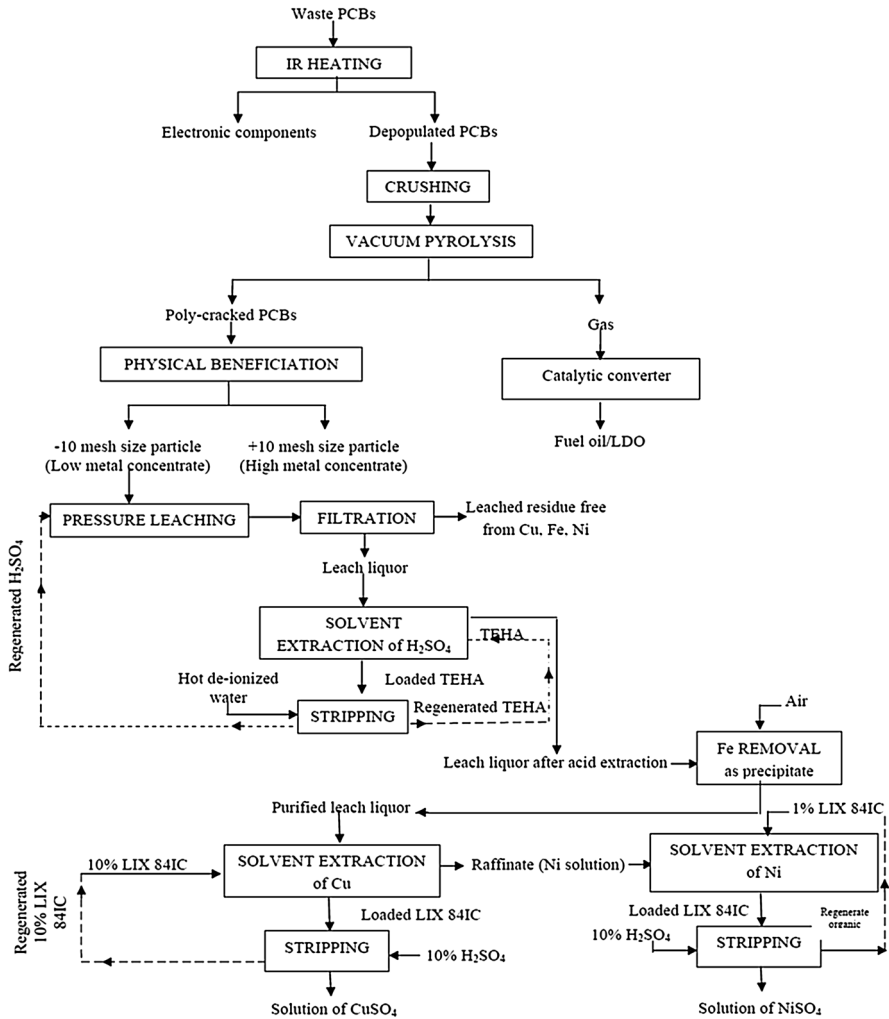


Fig. 5 Complete process flow sheets for recovery of base metals from waste PCBs [116]

oil it produced has unknown composition, which cannot be used for any refinery. Li et al. studied the vacuum pyrometallurgical process at low temperatures and concluded that vacuum (less than 1000 Pa) will significantly improve the performance of recycling by significantly reducing the activation energy from 127.87 and 115.36 kJ mol^{-1} to 53.59 kJ mol^{-1} , which significantly improves the outcome of the process [60].

However, the drawbacks of vacuum pyrometallurgy techniques are very obvious, namely, the requirement of a high vacuum. Although vacuum equipment is used in industry, the vacuum degree needed for the vacuum pyrometallurgy technique calls for expensive equipment and good air tightness, since the failure to guarantee high vacuum degree will lead to the incomplete pyrolysis of WPCB, causing hazardous

emissions and side reactions in the process. Therefore, the application of this process is seriously hindered due to its low economic feasibility.

Summarily, pyrometallurgy recycling techniques can recycle most of the MF at the cost of ruining the NMF. Also, the burn off of the nonmetallic fraction will not only produce bromorganic compounds and other toxic emissions, but it will also give around 20–25% ash content, which will have a certain percentage of heavy metals and need a further workload to perform disposal or refining [51]. A remediation method is to do the dehalogenation either no later than the pyrolysis process, or in the upgrading of the pyrolysis products [82, 98]. However, it may add further workload and cause the reduction of the yield and value of the pyrolysis products. Moreover, copper or lead in the feedstock will function as collectors of other metals, which will result in the output of unknown alloy from pyrometallurgy instead of pure metal that require further refining [99].

4.1.2 Hydrometallurgy recycling techniques

Hydrometallurgy recycling techniques make use of solvent leaching using cyanide, thiourea, thiosulfate, halide, and recycling the metallic fraction (mainly copper and gold) from WPCB feedstock [100–102]. Compared to pyrometallurgy, hydrometallurgy recycling techniques are more accurate, highly predictable, and easily controlled, which makes it the most competitive technique for WPCB recycling currently with the assistance of mechanical crushing as pretreatment [103].

Kinoshita et al. use nitric acid as a leaching reagent and they used two rounds of leaching in the first round Ni was extracted with a concentration of 279 mg/l at 363 K for 72 h and in the second round Cu was extracted with a concentration of 3220 mg/l at 363 K for 6 h [102]. The Au was obtained by filtration of the leachant and separated in the form of flakes. The metal-rich solution was forward extracted and backward extracted by organic solvent and 4.0 M nitric acid, respectively. The recovery rate is high and selective recovery was achieved. However, the process takes a long time and it produces waste water containing an organic phase and a strong acid, which is a common issue in hydrometallurgy recycling techniques. Similarly, Park et al. use aqua regia as the leaching agent to extract palladium, silver, and gold from WPCB feedstock [50]. The recovery of palladium in the form of $\text{Pd}(\text{NH}_4)_2\text{Cl}_6$ precipitation is 93 wt%. Gold nanoparticles were obtained by organic solvent extraction using dodecanethiol and sodium borohydride with a recovery rate of 97 wt%. Silver stays stable in aqua regia and was recovered without further treatment with a recovery rate of 98 wt%. The recovery rate is very high as well as the purity of the metal they collected. However, the usage of strong acid and organic solvent decrease the environmental evaluation of this process.

Therefore, despite the advantages of the hydrometallurgical treatment methods, a serious issues is that they produce a lot of highly toxic waste water containing cyanides or halides, which is extremely hazardous for both soil and water bodies [104]. Moreover, non-cyanide or non-halide leaching solvents like thiourea or thiosulfate have disadvantages including low-stability, high cost, and high consumption of extracting reagent [21–23]. Corrosion also causes problems for

Table 8 Comparison of different kinds of leaching solvent

Leaching solvent	Advantages	Disadvantages
Acid and mix acid [103–105]	High dissolution rate	Corrosive, high concentration of acid needed
Cyanide [106, 107]	Low cost, high stability, low dosage	High toxicity, slow leaching rate
Thiourea [53, 108, 109]	Low toxicity	Difficulty in recovery, high cost
Thiosulfate [110–112]	High selectivity, nontoxic, and non-corrosive	Sensitive to pH change
Halide [113]	High leaching rate, good selectivity	High cost, environmental irritation
Hydrogen peroxide [114, 115]	Low toxicity	Need acid assistance

the equipment when several kinds of leaching reagents were applied. More details are shown in Table 8 below.

Also, the leaching process normally takes a long time to obtain metal-rich solution due to the slow leaching rate, which makes the process a time-consuming one. Therefore, studies that reinforce the driving force and accelerate the leaching rate have been studied including using electrochemistry, pre-pyrolysis, supercritical extraction, and mechanical treatment [52, 54, 99, 117, 118]. Havlik et al. studied the hydrometallurgical process with pre-combustion, in which WPCB was leached in 1 M HCl solution after combustion in air for 15 to 60 min in 500–900 °C [99, 119]. The results showed that the pre combustion at 900 °C will significantly improve the leaching of copper from the WPCB feedstock due to the conversion of Cu to Cu₂O, which dissolves more preferable in HCl. However, the pre-combustion leads to the difficulty in extracting tin since Sn is oxidized to form SnO₂, which is very stable in acid with minimal leaching. Therefore, the efficiency of hydrometallurgy was seldom enhanced even without considering the emission during combustion process.

Fogarasi et al. used mediated electrochemical oxidation to recover copper from WPCB and high purity copper (99.04%) was obtained with 63.84% current efficiency [52]. However, a sludge was still produced with certain concentrations of copper and lead (49.37 and 32.49 wt%, respectively); this issue needs further work. Also, the fate of NMF was not clarified, as well as the bromide inside the NMF. Kim et al. use electro-generated chlorine to leach metals from WPCB feedstock [120, 121]. Copper as well as noble metal was obtained together due to the high oxidation potential they gained in the process. In the first stage, the recovery of Cu is 94.91% and in the second stage the recovery rate of Au is 93.06%. Then the metal ions can be collected by an ion-exchanger with efficiency around 97%. However, the feedstock used is already after dismantling and sorting therefore it has very high metal concentration (Cu 66 wt% and Au 0.045 wt%), which is not comparable to the raw WPCB feedstock, making the research not representative for all kinds of WPCB. Also, although the acid solutions as well as the low concentration solution are recycled back after ion exchange, the problem of discharge still exists since the halide is concentrated in the process if it is applied to WPCB with a high NMF content as well as the low pH of the solution. Silvas et al. used magnetic separation before the leaching test and they recovered 90 wt% Al, 40 wt% Zn, 8.6 wt% Sn in

the first leaching stage using sulfuric acid media and 100 wt% Cu, 60 wt% Zn, and 10 wt% Al in the second stage using oxidant [54]. It is supposed that the recovery rate is very high due to the assistance of mechanical separation and this could be a possible way for future work. However, the fate of NMF was not mentioned in this paper and remains a potential pollutant.

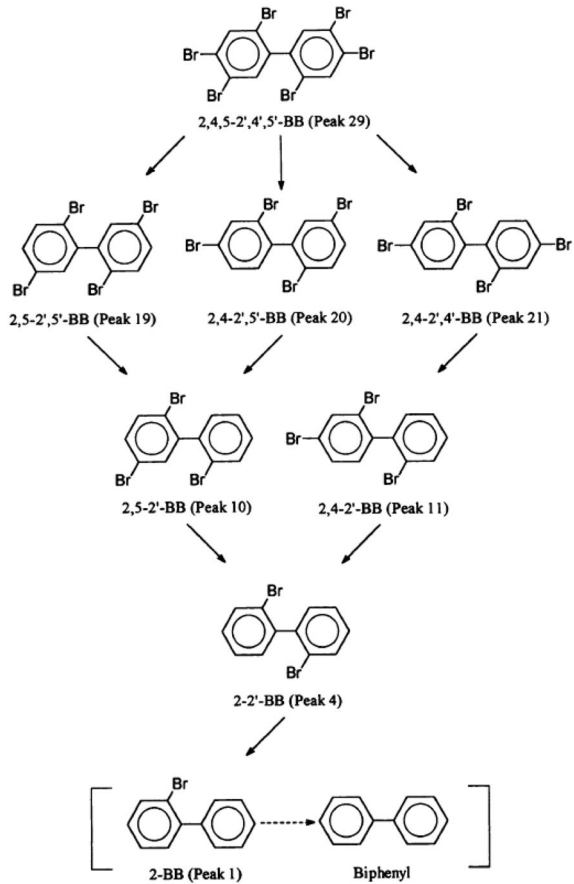
Summarily, hydrometallurgy recycling techniques are easy to apply and simple to operate. However, the problem that cannot be avoided or ignored is the discharge of leachate as well as the pollutants since these techniques do not incorporate the recycling of NMF. The process is mainly undermining the structure of NMF and emits them, which not only wastes the useful part of NMF but also converts them into pollutants. Another issue that should be highlighted is that the recovery rate for hydrometallurgy recycling techniques is full recovery. Therefore, there will be a certain amount of heavy metal present in the effluent in the form of free ions, which will reinforce the hazardous content of it. So an upgrading of current hydrometallurgical recycling techniques is urgently required to stop the wild application of it.

4.1.3 Biometallurgy recycling techniques

Biometallurgy recycling techniques use microorganisms including bacteria or fungi to treat WPCB [122]. Commonly used bacteria include mesophilic chemolithotrophic, cyanogenic or moderately thermophilic bacteria [25, 26, 123, 124]. The mechanism of biometallurgy recycling techniques is similar to hydrometallurgy recycling techniques since they all incorporate the process of leaching. However, instead of adding leaching reagents, biometallurgy recycling techniques normally use the chemicals produced by the microorganism itself, including organic/inorganic acids, cyanide, or sulfate ions. After leaching, the metal ions will form complexes or precipitates and thus they are separated from the culture broth for direct use or further refining. It has the advantages of only a small volume of waste water discharge and is environmental benign compared to hydrometallurgy, which generally requires a high dosage of toxic chemical reagents. Besides, certain kinds of bacteria are capable of reducing brominated flame retardants in the pathway shown in Fig. 6, which is rarely achieved in other recycling techniques [122, 125].

Liang et al. used a mixed culture of *Acidithiobacillus thiooxidans* and *Acidithiobacillus ferrooxidans* to recover copper from WPCB fine powder [123]. The bacteria can oxidize elemental sulfur added to sulfuric acid for bioleaching and then extract the metal from the broth. The highest recovery rate (98.37%) was obtained with the condition of pH value 1.56, elemental sulfur S^0 5.44 g/l and 16.88 g/l $FeSO_4 \cdot 7H_2O$ concentration. However, the increase of WPCB addition in culture from 18 to 32.4 g/l will cause a sharp drop in the copper recovery rate from 98.3 to 87.2%. Also, the fate of NMF was not mentioned in this work and remains a potential hazardous material. Ilyas et al. used moderately thermophilic bacteria to recover metals and conducted the process in a column test [124]. The recovery rate for Zn, Al, Cu, and Ni is 80, 64, 86, and 74%, respectively, which already meets the requirements of industrial-scale implementation for recycling of MF of WPCB. It is also noticed that NMF will contribute to the alkalinity which will affect the leaching of metal ions, requiring washing before conducting the culture stage. However, in

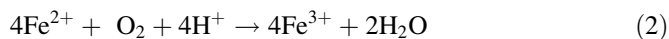
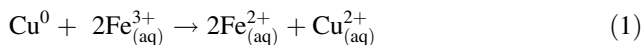
Fig. 6 Proposed pathway for the reductive debromination of 2,4,5-2',4',5'-BB in Firemaster by Pine River and Hudson River sediment microorganisms. Debromination of 2,2'-BB was only observed for a pyruvate plus Aroclor 1242 enrichment obtained from Hudson River sediment microorganisms



this process, Pb and Sn along with other metals will form precipitates in the column, not only preventing the recycling of metals but also causing potential blockages of the column. Yang et al. used *A. ferrooxidans* to study the factors affecting the mobilization of copper in the bioleaching process [125]. The higher concentration of Fe^{3+} from 0.64 to 2.13 g/l in the stock solution will bring to the increase of copper leaching in 12 h from 34.53 to 79.75%. As a similar result was observed that copper recovery decreased from 99.06 to 88.40% in 48 h when the pH value increased from 1.5 to 2.0. Therefore, it is noticed that the concentration of Fe^{3+} as well as the pH value have a very obvious effect on copper leaching. Ting et al. used two cyanide-producing bacteria, *Pseudomonas fluorescens* and *Chromobacterium violaceum* to extract gold and copper from WPCB and the recovery rate was around 27 and 20% for gold and copper [126]. In a two-step extraction, the recovery rate for gold and copper was increased to around 30 and 24%, respectively.

However, biometallurgy recycling techniques require a lot of nutrients for microorganism enrichment and metal extraction. The addition of an Fe source is necessary for copper extraction since the solubilization of copper needs the presence of Fe^{3+} according to reaction (1). Also, the ferrous ion is the energy source for *A.*

ferrooxidans, which is an aerobic and autotrophic bacterium according to reaction (2).



Also, the low extraction rate in high WPCB dosage due to the limitation in air distribution and oxygen mass transfer hinder the application of the biometallurgical treatment method [127]. Moreover, normally microorganisms are vulnerable to heavy metals, thus the growth of them will be inhibited due to the toxicity of metals [128]. Although some bacteria or fungi can adjust to the condition after prolonged adaptation time and achieve a good recovery rate, the time required for this adaption is extremely long (more than a week) [129]. Furthermore, the recovery period for biometallurgical treatment is much longer than pyrometallurgy or hydrometallurgy recycling techniques, which affected the positive evaluation of process. Also, normally the WPCB feedstock needs to be a fine powder with a particle size around 100 μm or even lower to ensure adequate surface contact, which will consume a lot of energy in the early stage [130].

4.2 Summary of partial recovery of metallic fraction from WPCB

From the above-mentioned techniques, partial recovery of the metallic fraction from WPCB processes will benefit metal recovery, which incorporates the most valuable components inside WPCB. The application of partial recovery of metallic fraction from WPCB still exists in many developing countries trapped by the fact of lacking funding. Differences still exist among pyrometallurgy, hydrometallurgy, and biometallurgy in terms of cost, efficiency, harmful effect, as well as the time for recycling as shown in Table 9.

Pyrometallurgy shows the best performance in efficiency and time for recycling since it can significantly remove the non-metallic fraction in WPCB by thermo-decomposition. However, the high energy consumption and construction cost also make it the most expensive technique. Additionally, the toxic emissions and residues generated in the process cause considerable harmful effects to the environment if performed without proper treatment. Compared to pyrometallurgy, hydrometallurgy and biometallurgy have much less environmental impact since the pollution is confined mostly in the liquid phase. Nevertheless, the driving force in hydrometallurgy and biometallurgy is not comparable to pyrometallurgy, thus making them extremely time-consuming techniques although their cost is several times lower than pyrometallurgy. Summarily, the partial recovery of metallic fraction from WPCB by the above methods has obvious drawbacks. It is imperative to develop further full recovery techniques for WPCB to improve the current methods of WPCB recycling.

Table 9 Comparison of different kinds of partial recovery of metallic fraction techniques

Technique	Cost	Efficiency	Harmful effect	Time for recycling
Pyrometallurgy	High	High	High	Short
Hydrometallurgy	Low	Medium	Medium	Medium
Biometallurgy	Medium	Low	Low	Long

4.3 Primitive full recovery from WPCB

Despite diversity possessed by MF recycling techniques, a common drawback shared by them is the absence of NMF treatment, which will give rise to the toxic emission including brominated organic compounds, secondary particulates, and other kinds of pollutants. A simple mass balance concept implies the less recycled, the more emitted. Therefore, techniques focusing not only recycling the MF but also achieving the safe disposal of NMF simultaneously are reviewed next including supercritical fluid extraction, plasma treatment and hydrothermal method [131–133].

Huang et al. used DC arc plasma to treat printed circuit boards, which decomposed the WPCB in molten bath at 1400–1800 °C in a DC arc furnace [131]. The product for this process is homogenous and vitreous slag and small molecular gases including HCl, H₂S, NO_x, and SO₂. Although the safe disposal of solid residue is achieved, it produces a certain amount of air pollution and the energy consumption is huge due to the high temperature and long duration time. Xiu et al. has conducted a series of studies using supercritical fluid including supercritical water (SCW) and supercritical methanol (SCM) to recycle WPCB [134–138]. For the SCM recycling, most kinds of heavy metals are converted to metal oxides with around 100% conversion, except for Mn and Ni, since near 50% of them enters in liquid phase. The copper extracted from WPCB will undergo an eletrokinetic (EK) process and can be synthesized to nanoparticles, which functions as a photocatalyst [139]. The degradation of NMF also happens with the formation of phenol and phenol derivations, which is the main composition of liquid product (~55%). Xing et al. also used SCW to detoxify bromorganic compounds inside WPCB and achieved maximum debromination rate of 97.8% [55, 140]. The bromide was concentrated in water in the form of hydrogen bromide. Also, the bromine-free oil with main components consisting of phenol (58.5%) and 4-(1-methylethyl)-phenol (21.7%) was collected. Moreover, copper was recovered in the purities of near 95% with a recovery rate as high as 98.11%. However, supercritical technologies need high temperature (above 400 °C) and high pressure (higher than 20 MPa). Therefore, the utilization of this process needs a good-quality equipment, which increases the cost significantly. Yin et al. used hydrothermal technology to decompose brominated epoxy resin in WPCB and achieved more than 80% decomposition rate and obtained a liquid with the main composition of phenol, which can be used as a chemical material [141]. Also, after this treatment, WPCB can be used to recover MF with a much lower emission. However, the feature of hydrothermal technology determines that the operation capacity is very limited; in this work, 0.1 g WPCB per batch, which is not applicable for large-scale operations.

Summarily, despite the fact that the previously mentioned technology can recycle MF with the safe disposal of NMF, the energy and effort put into this process is not proportional to the output, which is only the MF. Also, although the safe disposal of NMF will not cause further pollution, a considerable amount of resource inside NMF is wasted after all. Therefore, the separation of MF and NMF and further recycling of NMF is necessary for the consideration of sustainable resource recovery. For this, the effective separation for MF and NMF is indispensable for the purpose of more detailed and efficient recycling.

5 Advanced Recycling Techniques

5.1 Mechanic Separation of MF and NMF

Based on the previous Sects. 1, 2, 3, 4, it can be concluded that a common drawback shared by the primitive recycling technologies is the lack of attention given to the NMF-70% by weight of the WPCB, which fundamentally blocks the recycling or upgrading of NMF due to the loss of mechanical or chemical structure and properties. Therefore, separation of MF and NMF without damage to the structure of NMF is the precondition for NMF recycling and itself is also regarded as one type of recycling since the MF separated from the feedstock can be sold to the market directly. With the development of technologies, the method for MF and NMF separation has changed from manual disassembly to more advanced technologies. Mechanical separation methods take the advantages of the differences in density, magnetism, and electro-conductivity, and have become increasingly important in the recycling of WPCB [27–30]. Since the composition of NMF indicates that most chemical separation techniques will damage the properties of NMF and bring difficulties to further treatment operations, mechanical separations stand out although in some situations they were regarded as assistance processes for pyrometallurgy or hydrometallurgy techniques. Still it is an efficient recycling method which offers great potential, especially with the development of electrical conductivity-based separation [53]. Also, with the development of the automation industry, the mechanical separation also involves automatic or semi-automatic application [11]. These techniques can relieve the situation a lot by separating the MF and NMF of WPCB with a high efficiency and good purity. Therefore, it makes it possible to treat MF and NMF separately and it brings about the selling of high-purity MF to the market and also directing recycling and modification of NMF from WPCB as two potential ways to treat the NMF part of WPCB. Also, the separation of MF and NMF avoid potential interactions of NMF in further treatment stages because the metals in MF can catalyze unwanted side reactions of NMF.

5.2 Density-based Separation

Density-based separation uses the differences of particles size and density to separate the MF and NMF and various equipment items were developed, as shown in Fig. 7 [27]. This method has a long history dating back to the jigging method,

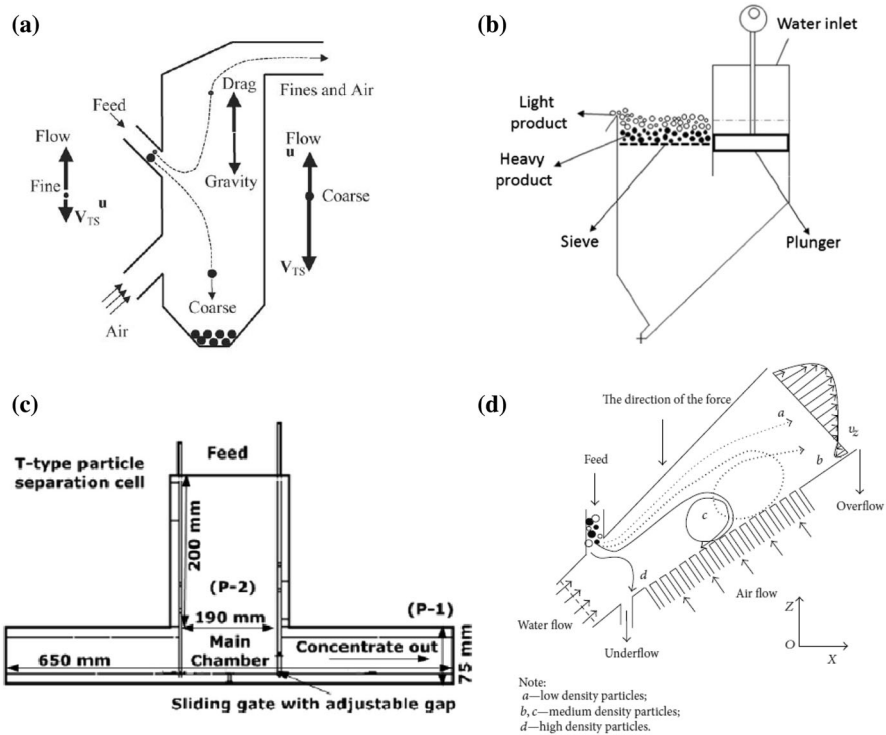


Fig. 7 Schematic illustration of several kinds of density-based separation methods: **a** air classifier, **b** laboratory scale Harz jig, **c** T-type vertical vibration separation cell, and **d** inflatable tapered diameter separation bed

which used a plunger to hit the bottom of the feedstock to give a force to kick the light particles out, thus preserving the heavier ones. Sarvar et al. used wet jigging to separate WPCB feedstock and the recovery rate of metal content for coarse-size, middle-size, and small-size fractions are 95.6, 97.5, and 85%, respectively [142]. However, low grades around 70% were obtained and a significant part of gold is lost at the flotation process. Currently, air classification is a popular method based on the fact that the particles suspended in the gas stream will be separated due to the gravity and drag forces experiencing in opposite directions [28]. The heavier particles will gain larger terminal settling velocity and move towards the bottom of air steam while light particles go to the opposite side, which is the top of the column. Eswaraiah et al. used an air classifier to separate the WPCB and recover 96.7% copper in the sink and 98% plastics and glass fibers in the float [143]. However, there is still 70.94% of other materials in the sink, which makes the purity of copper only 26.77%. Habib et al. [144] also use vertical vibration to separate MF and NMF from WPCB feedstock and they obtained a MF fraction as a combination of Cu (~50%), Fe(~10%), Sn(~10%), Zn(~8%), Pb(~8%), as well as some NMF. Also, the NMF fraction includes 65% NMF and 25% Cu as well as a certain amount of metals. The advantages of density-based separation are the simplicity of

its equipment and low energy consumption while the common problem is low product purity, adding workload and difficulty to further refine [145].

5.3 Magnetic-based separation

Magnetic-based separations can achieve the recovery of ferromagnetic metals such as aluminum from non-ferrous metals and other non-magnetic contents with a permanent or electro-magnet. An eddy current separator is a mature technology for magnetic-based separation that can efficiently separate aluminum from non-ferrous contents, as shown in Fig. 8 [29]. When the feedstock passes over the separator, aluminum will be affected by the eddy current generated by the rotation of the magnets inside the shell at high speed, which causes them to be surrounded by a magnetic field. Therefore, the particles will be repelled away from the magnet much further due to the polarity between the two magnetic fields, thus the separation between them and NMF is achieved. Zhang et al. used an eddy current separator to recovery aluminum from electronic scrap and achieved more than 90% aluminum recovery rate with a purity around 85% for a single pass [146]. Yoo et al. used a two-step magnetic separation to recover MF from WPCB and from the first magnetic separation at 700 Gauss, 83% of the nickel and iron was recovered in the magnetic fraction. The second magnetic separation at 3000 Gauss increased the total amount of nickel recovery but caused a drop of the nickel purity from 76 to 56% [58].

However, not all MF can be separated out through eddy current separation because only material with a high σ/ρ can be separated out, where ρ is the density of the material and σ is the electrical conductivity of material. Table 10 shows the materials that can be separated by an eddy current separator, from which it is obvious that aluminum is the most easily separated material while stainless-steel, plastic, and glass have a zero value for the conductivity-to-density ratio, meaning it is not applicable to separate them by an eddy current separator [147].

5.4 Electronic conductivity-based separation

Electronic conductivity-based separation focuses on the difference in the electrical conductivity between MF and NMF, which is normally related to corona

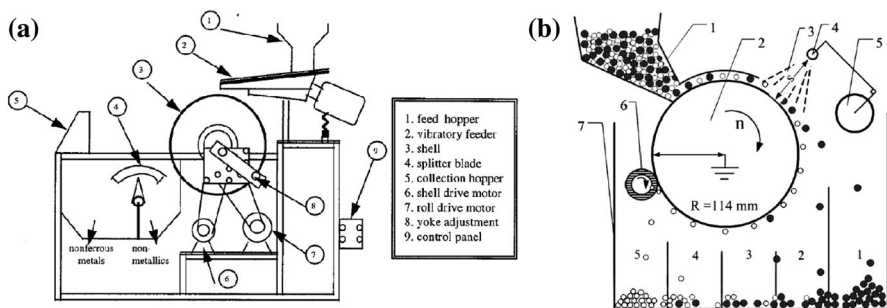


Fig. 8 Schematic illustration of eddy current separator (a) and corona electrostatic separator (b)

Table 10 Materials that can be separated by an eddy current separator, and their properties

Materials	σ ($10^{-8}/\Omega\text{m}$)	ρ (10^3 kg/m^3)	σ/ρ ($10^3 \text{ m}^2/\Omega\text{kg}$)
Al	0.35	2.7	13.1
Zn	0.17	7.1	2.4
Ag	0.63	10.5	6
Cu	0.59	8.9	6.6
Brass	0.14	8.5	1.7
Pb	0.05	11.3	0.4

σ electrical conductivity, ρ density, σ/ρ ratio of electrical conductivity to density

electrostatic separation in PCB recycling that fits the condition well, as shown in Fig. 8 [30]. The sample feed in the separator will be bombarded by the high-voltage electrostatic field generated by a corona electrode and electrostatic electrode. Now, the MF will be neutralized quickly as they contact the earthed electrode and leave the rotating roller while the charged NMF are pinned by the electric image force to the rotating roller and move with the rotating roller, finally falling into the holding tanks.

Li et al. used corona electrostatic separation with the pretreatment of pulverization of WPCB feedstock to successfully separate the MF and NMF of WPCB and achieved a high recovery rate (more than 90%) along with high capacity (0.5-1 t/h) with no obvious side effects compared to fluidized bed separation, which has wastewater discharge or air-current separation with a dust-releasing issue [19, 148]. The MF has very high purity and can be sent to a smelting plant directly or with minimal refining. Jiang et al. has designed a new two-roll electrostatic separator that takes advantage of the force of gravity to pass the mixture to the second step for recycling of metals and nonmetals from waste printed circuit board [149]. The production capacity was significantly increased for maximum 50% with 45% reduction of middling products. However, mechanical processes for recycling WPCB normally needs to undergo at least two steps, including coarse crushing and fine pulverizing and especially during fine pulverizing, the temperature will increase up to almost 300 °C [150]. Dust and ash is a common issue for mechanical separation and although it can be avoided by using personnel protective equipments, it still adds to the cost of the process, which makes the economical feasibility of this energy-intensive process even worse. Therefore, an upgrading of NMF to make value-added products is necessary to remedy the expenditure of energy and equipments cost. Also, noble metals will be lost in the crushing process since they will attach on the surface of nonmetallic fraction and cannot be recycled [49].

5.5 Directly Recycling of Nonmetallic Fraction from WPCB

At the beginning, the purpose of recycling of NMF from WPCB is simply to avert the potential hazardous environmental impacts when they end up in landfill or incineration. The composition of NMF is a random combination of epoxy resin and glass fiber. The reuse of NMF is hindered due to understanding of the chemistry of them. Therefore, it is common to be directly used as filler, concrete, and modifier,

which means the NMF was used without any forms of modification and is a low value-added material [31–35, 151–154].

Guo et al. has done a series of studies on the feasibility of replacing wood flour in the production of phenolic moulding compound (PMC) using the NMF of WPCB [151, 152]. The addition of NMF can significantly improve the impact strength and heat deflection temperature (HDT) and reduces flexural strength and Raschig fluidity of the phenolic moulding. Notably, the increase of the content of NMF will sharply reduce Raschig fluidity. Therefore, the optimum content for adding NMF is considered to be 20 wt%, in which condition the flexural strength is 70 MPa, the Charpy notched impact strength is 2.4 kJ/m², the heat of deflection temperature is 168 °C, the dielectric strength is 3.9 MV/m and a Raschig fluidity of 103 mm. These criteria all meet the national standard. In addition, they did economic analysis, which shows that the addition of NMF can save the cost for the producers of PMC. However, if the cost-saving can cover the energy consumption of corona electrostatic separation is not clear. The authors claim the market price of NMF is for free, which is not correct in the current market. They also study the possibility of producing nonmetallic plate (NMP) from NMF of pulverized WPCBs by hot-press forming with the addition of a resin paste as a bonding agent [31, 32]. The maximum content of NMF in reproduction nonmetallic plate (RNMP) can be as high as 40 wt%. However, the RNMP showed excellent mechanical properties with impact strength of 5.8 kJ/m² and flexural strength of 65.1 MPa when NMF added was 20 wt%. Yokoyama and Iji also investigated the use of NMF as filler in the reproduction of resin-type construction materials and the comparison of mechanical properties of these materials with those of reference materials with silica powder was conducted [33]. The NMFs in the materials show reinforcement in mechanical strength and thermal expansion properties of the epoxy resin mold, which has better performance than talc, calcium carbonate, and silica. This is probably because of the compatibility between the NMFs and the epoxy resin matrix, and also the incorporation of glass fiber.

Niu and Li used recycled waste PCBs for cement solidification, which is actually a method to use the waste PCBs as a raw material for concrete [154]. It is proved that the cement solidification can be significantly improved in terms of the compressive strengths (4.89 MPa for slag cement and 7.93 MPa for Portland cement) and the impact resistance of 200 (maximum) of NMF can turn it into strong monoliths. The leaching test shows that the leaching of Pb in the raw material can be effectively prevented (<5 mg/l) even under an acidic environment.

Zheng et al. used NMFs as reinforcing fillers in PP composites [153]. The NMFs are modified by a silane-coupling agent KH-550 and PP powder S1003 is applied as the matrix polymer. NMF and PP powder are pre-mixed and the products are obtained by extruding. The mechanical characterization shows that the ensile strength, tensile modulus, flexural strength, and flexural modulus are significantly improved 28.4, 62.9, 87.8, and 133.0%, respectively, by the adding of NMF. The optimum content of the NMF added is 30 wt% based on technical, environmental, and economical consideration.

However, Lu et al. found that PCB can be liberated effectively with a size between 0.5 and 1.2 mm while for a better recovery ratio the over-pulverized

phenomenon is very serious. This limits the further use of the glass fibers that are the support material of PCB (~50 wt%) [155]. The mechanical properties of NMF were sacrificed in order to achieve a high metal recovery rate. Therefore, the direct reuse of NMF is seriously influenced due to this phenomenon. A more advanced technology of NMF modification is indispensable.

5.6 Modification of Nonmetallic Fraction from WPCB

The direct recycling of NMF can help solve the disposal problem of NMF. However, it is still weak for them to compensate for the high operation cost of the MF and NMF separation due to the low value-added products they produced by NMF. With the requirement to better use NMF, which takes more than half of the weight of NMF, the modification of NMF followed by the upgrading was studied by many research groups [156–160].

One thought for the modification is based on the thought that NMF is a carbon source for potential reuse considering the high content of carbon in its composition due to the addition of epoxy resin or other polymers. Normally, the carbon content in NMF is between 30 and 40%, which varies with the source of WPCB [161]. Ke et al. first pyrolyzed the NMF of WPCB in the temperature range 500–800 °C and used physical or chemical activation to obtain activated carbon [156]. Physical activation with H₂O as an activation reagent produced granular activated carbon with a surface area as high as 1019 m²/g and pore volume 1.1 cm³/g while chemical activation with KOH as activation reagent obtain the same product with higher surface area of 3112 m²/g and a pore volume of 1.13 cm³/g.

Rajagopal et al. used the activated carbon prepared by physical activation with CO₂ subsequent to pyrolysis of NMF of WPCB to and apply it in supercapacitor [157]. The NMF activated at 850°C for 5 h shows the highest surface area of 700 m²/g as well as 0.022 cm³/g pore volume. Electrochemical characterization of the activated carbon prepared under optimum conditions, shows a specific capacitance of 220 F/g at the current density of 30 mV/s and 156 F/g at the current density of 100 mV/s, which is comparable to activated carbon prepared by other methods. Also, the activated carbon has an energy density of 15.84 Wh/kg at 850 W/kg, which is very high compared to commercially available supercapacitors based on activated carbons that have an energy density ranging from 4 to 5 Wh/kg with power density values of 1 to 2 kW/kg [162]. Moreover, the retention value of the activated carbon was studied to be 98% for over 1000 cycles.

Hadi et al. has developed a thermal-alkaline activation process to functionalize NMF to produce an aluminosilicate adsorbent for heavy metals uptake from waste water [158]. The NMF was mixed with potassium hydroxide solution and activated in 300 °C to develop the porosity of NMF which is a non-porous material. After activation, equilibrium isothermal adsorption tests show that the modified novel material called ANMF had a high uptake capacity for Cu(2.9 mmol/g), Pb(3.4 mmol/g), Zn(2.0 mmol/g), which is much higher than commercial adsorbents used in industry. Xu et al. studied the factors affecting the adsorption capacity including contact time, initial cadmium ion concentration, pH, and adsorbent dosage when this material is used for cadmium uptake [159]. The results showed that pH

has an important effect on the uptake capacity for cadmium and the maximum uptake capacity for cadmium is 2.1 mmol/g obtained when $\text{pH} = 4$. Modeling of the adsorption was also done in this work and several isothermal equations have been studied while the Redlich–Peterson model shows the best fitting. Moreover, the activation mechanism as well as the optimization of reaction condition was done by Ning et al. [160]. The factors affecting the pore-development process were studied including reaction temperature, impregnation ratio, and reaction time, and the results shows that reaction temperature and impregnation ratio has a significant effect on the structure of ANMF during the reaction while reaction time has no significant effect. This provides a possibility for the porous structure tuning for the NMF of WPCB, which will significantly enlarge the potential applications of NMF as a catalyst, adsorbent, and filter support.

Summarily, the modification of NMF to make value-added products can greatly improve the value of NMF. Thus, the value added to NMF can possibly compensate for the high cost derived from mechanical separation.

6 Perspectives on Recycling Technologies

6.1 Economy Perspective

The economy perspective is always very important, or of first priority for the evaluation and implementation of a recycling technique. The process, which is cost-effective, will have a better chance to be commercialized especially in developing countries due to the lack of sufficient funds for WPCB recycling. Obviously, landfill and incineration has limited economical benefit even without the concern about the treatment of the hazardous substances they emit. The construction and administration costs of landfill sites or incineration plants are large expenses with a scant return. This is also a reason for the reduction in the current application of these two techniques for WPCB treatment. Pyrometallurgy and hydrometallurgy recycling techniques can achieve partial recycling of WPCB by recovery of MF from the feedstock. However, the intensive energy consumption in pyrometallurgy is ascribed for the main expense in the whole process. The energy power in the process is around 0.2–4 kW, varying with the different processes, while sufficient data is not available for these techniques since most works were conducted at the laboratory scale and energy analysis is absent for most of the studies [148]. However, the normal pyrolysis temperatures are from 400 °C to 800 °C, and the post gas treatment equipment normally needs the temperature higher than 1200 °C and no doubt consumes a considerable amount of energy. For hydrometallurgy recycling techniques, the chemical reagents used including cyanide, halide, acid, or other chemicals takes a considerable and indispensable percentage of the cost. Also, in hydrometallurgy, water is used as leaching media and solvent on a large scale, which also contributed to the operation cost.

Furthermore, as the decrease of percentage of noble metal in the WPCB is due to manufacturing upgrading, the driving force for recycling MF from WPCB is decreasing, since previous works focus on the benefit of metal recovery. Recycling

of NMF is a potential way to remedy the cost on energy and/or chemical cost. However, the precondition for NMF recycling is the homogenization of the WPCB, which means the pulverization of WPCB feedstock by shredders and hammer mills, which also requires energy and equipment cost. A fine size of NMF (around 0.1 mm) is typical for the recycling of NMF in many studies. While in both pyrometallurgy and hydrometallurgy recycling techniques, the structure of NMF was totally decomposed, the recycling of NMF is therefore not applicable. In some types of mechanical separation, the structure or composition of NMF was preserved with a meager effect on the chemical and mechanical properties. Nevertheless, the direct use is not enough to add adequate value to compensate for the cost required to preserve their structure. Therefore, the upgrading and recycling of the value-added non-metallic fraction are attracting more and more researchers.

6.2 Environmental Perspective

The environmental perspective is another important issue that affects the evaluation of the WPCB recycling techniques. A general rule for emissions is that the more material recycled, the less will be emitted. Therefore, the focus of a technique should not only be on the performance of the recovery rate of specific components like copper or noble metals, but also the fate of the remaining part due to the potential emissions they may have, especially for gaseous emissions. The environmental emissions can be divided into primary pollution, which comes from the WPCB, and secondary pollution, which is emitted during the treatment or recycling process.

For primary pollution, heavy metals including Cu, Pb, Cd are of great concern. They can enter the environment in the form of leachate in landfill or vapor in incineration or some pyrometallurgy recycling techniques without proper purification. It can cause the distortion of the human body as well as various other hazardous effects; the presence of PBDD/Fs and PCDD/Fs is a critical health issue when their presence in the vapor will cause cancer. They mainly form due to the oxidizing atmosphere and the presence of metals as catalyst.

However, the majority of public attention is still paid to the emissions of NO_x, SO_x, or VOCs, while some kinds of pollutants are ignored due to their relatively low toxicity, like Br and HBr. Yet low toxicity does not equal being environmentally friendly. The vast emissions still cause damage to individual hygiene [19]. Bromide is a hazardous material source with obviously inadequate attention, it is even claimed as harmless in a few studies [5, 15]. Bromide concentration in WPCB is relatively high (~ 4%) due to the addition of brominated flame retardants [163]. The decomposition of brominated flame retardants happens in the recycling process while bromide still remains in the form of several kinds of bromide or bromorganic compounds including HBr, bromomethane, and bromophenol (Fig. 9). However, research shows that bromide, especially HBr, has a very bad effect on human health, since it can cause health problems and is accumulated in the human body and is difficult to degrade. Therefore, to achieve complete recycling and elimination, emissions are the ultimate objective of the environmental impact of WPCB recycling techniques.

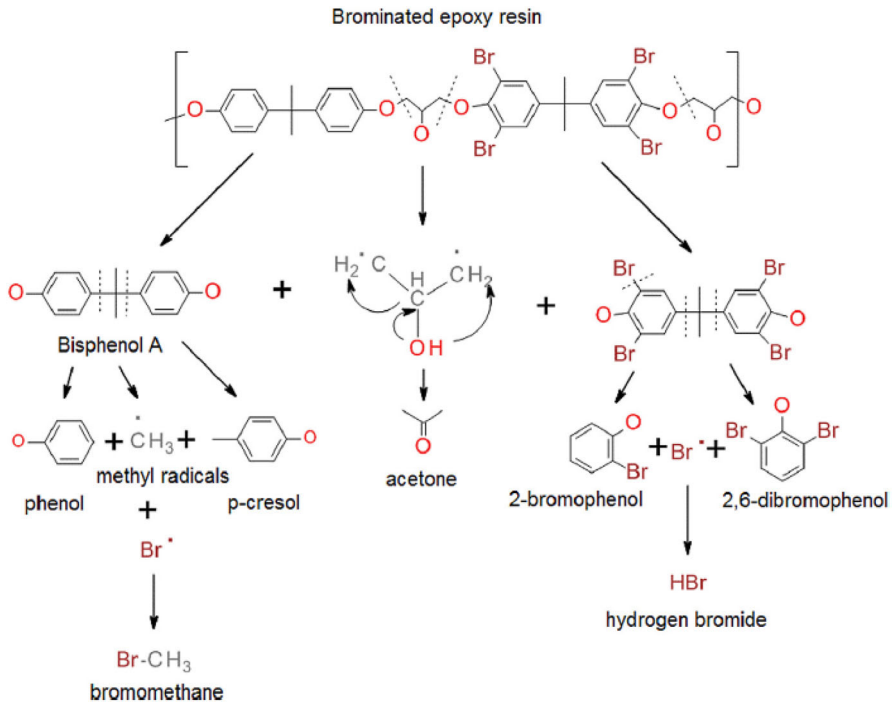


Fig. 9 Possible decomposition pathway of brominated epoxy resin according to the products generated [51]

6.3 Gate-to-Market Ability Perspective

Previously, the evaluation of a recycling technique is always based on economic and environmental perspectives. However, for a real application, it is not a two-sided choice while sometimes a technique is both cost-effective and environmentally friendly and does not mean that this process has high feasibility since the product's status is not well clarified. In previous research, a common mentioned concept is the recovery rate, which means the percentage of materials recovered from feedstock, in weight or volume percentage. Yet with deeper understanding and the real need for resources, the recovery rate is not comprehensive enough to describe the extent of recycling due to the fact that it is not capable of reflecting the required purity of the products, as for some techniques the recovery rate is very high (>90%) while the product purity is particularly low, including several components with concentrations between 20 and 40%. These products are actually incapable of being used or sold to the market directly, which need further separation or refining. However, in the evaluation of the process, the cost and environmental effects are not ascribed to the technique itself but are transferred to the uncertain downstream treatment. Thus, the performance of the technology can be overestimated or underestimated. Therefore, in this work, a new criteria was first raised named "gate-to-market ability", which means the ability of the products to be sold or used in a real industry application

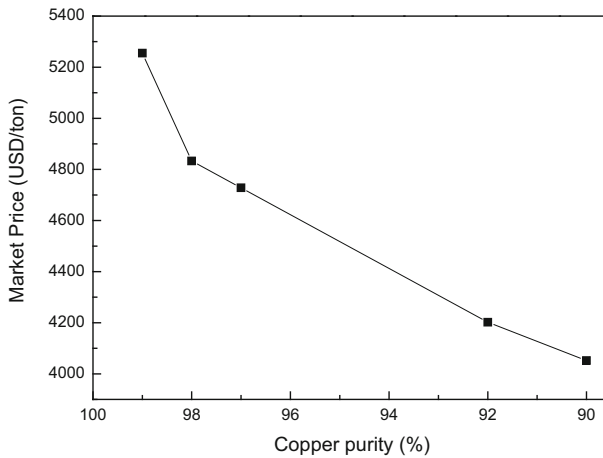


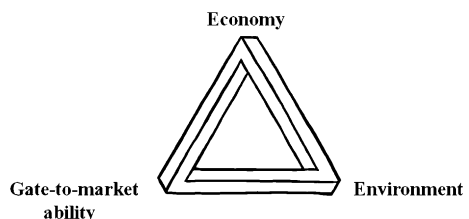
Fig. 10 Market price of copper with different purity (collected on January 21, 2016)

after it has been produced by the technique. It is obvious that the purity of the products will be significantly affected by their gate-to-market ability of them, for some recycling produces even a slightly lower purity will cause them to be discarded.

The market price of copper, a common recycling product of WPCB, is seriously affected by their purity, as shown in Fig. 10 where the prices of copper were collected from several copper markets over the world on January 21, 2016. It can be concluded from the figure that a small decrease in purity (from 99 to 90%) will cause a sharp drop in the unit price of around 22.89% when the purity is decreased. However, the copper recovered from the above-mentioned technology is normally around 70%, except corona electrostatic separation, which means the margin of profit is cut a lot or even not applicable without further refinery. The following process will add both an economic and environmental burden to the techniques. Therefore, gate-to-market ability is a criterion independent of economical and environmental perspective and plays an important role in the evolution of the feasibility of the process.

Normally, hydrometallurgy recycling techniques will give low gate-to-market ability due to the form of products, which are normally alloys. Generally, hydrometallurgy recycling techniques and mechanical separation can provide products with good gate-to-market ability since the products of these processes can

Fig. 11 The relationship for three factors on technology evaluation. This triangle actually does not exist, indicating the impossibility of achieving these three criteria simultaneously



achieve high purity beyond 90%, which reduces the difficulty of further application. Still, co-precipitation in hydrometallurgy will reduce the gate-to-market ability since the product will have several kinds of components inside.

7 Conclusions

WPCB is dangerous waste, but at the same time a rich resource for various kinds of materials. The recycling technologies of WPCB divided into direct treatment (landfill and incineration), primitive recycling technology (pyrometallurgy, hydrometallurgy, biometallurgy and primitive full recovery of NMF), and advanced recycling technology (mechanic separation, directly use and modification of NMF) were studied and analyzed to access their advantages and disadvantages. This review shows that direct treatment has advantages on their simplicity of treatment but the environmental problem is a big issue for them. Primitive recycling techniques achieve a certain extent of recycling, mainly the MF of WPCB. However, the energy consumption as well as the waste gas and water emission still remain a problem for the application of them. Advanced recycling techniques can separate the MF and NMF of WPCB with minimum effects on the environment with a good gate-to-market ability while the energy consumption is huge for them. Therefore, the direct use and modification of NMF was studied to remedy the cost from the energy consumption and become the concern of WPCB recycling.

To summarize, a triangle figure of economy perspective, environmental perspective, and gate-to-market ability perspective can be set as shown in Fig. 11. However, the discussion indicates that the three criteria cannot be satisfied with one simple technique. Therefore, the future research direction of WPCB recycling should focus on the combination of several techniques or in series recycling since the drawback of a process could have a chance to be remedied by one of the techniques and the evaluation should be conducted on the whole process until the products satisfy the market standard.

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